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Behaviour of dielectric barrier discharges in nitrogen / oxygen mixtures

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Dielectric barrier discharges have been investigated in binary mixtures of nitrogen and oxygen by means of spatio-temporally resolved optical emission spectroscopy. The content of oxygen was varied within the range 0.5-97%. Qualitatively, no profound influence of the gas composition on the discharge behaviour has been found. Observed quantitative changes in the luminosity distributions depending on the oxygen content in a mixture, may be explained by the role of collisional quenching of nitrogen excited states by the O_2 molecules.

1. Introduction

Barrier discharges (BDs) in oxygen-containing gas mixtures consist of many tiny filaments (microdischarges - MDs) of nanosecond duration. Due to their small dimensions (typically, a few mm) and short lifetimes, there is a lack of experimental data related to their spatial structure and evolution, that are needed to provide an adequate quantitative theoretical description for the MD development. To fill this lag in understanding the mechanism of BDs (MDs), systematic investigation for the case of binary gas mixtures (N_2+O_2) of variable composition have been undertaken.

2. Experimental set-up

The BD was generated in a flowing gas mixture of nitrogen and oxygen in a discharge cell consisting of two semi-spherical electrodes, both covered by glass (gap width d = 1.4 mm). The content of oxygen was varied in the range of 0.5-97 %. An applied sinusoidal voltage (frequency f = 6.5 kHz) with amplitudes U(peak-to-peak) = 16-19 kV was used to sustain the discharge with 1-3 MDs per half cycle. The BD was investigated by current oscillography, optical emission spectroscopy, and cross-correlation spectroscopy (CCS) for spatio-temporally resolved measurements of the MD evolution. A detailed description of the experimental set-up and the CCS method is given in [1]. Therefore the most important measurement characteristics are summarised: the spatial resolution along discharge axis is 0.1 mm, while 0.3 mm in radial direction, the wavelength accuracy is 0.3 nm, the resolution over the fine time scale is 0.1 ns, and additionally the measurements are resolved over the phase of driving sinusoidal voltage with T/16 (T = 1/6.5 kHz = 153 μs).

3. Results and discussion

In pure nitrogen, a diffuse ("glow", Townsend-like) BD is generated [2]. But an admixture of even some hundred ppm of oxygen leads to the transition to the filamentary mode, characterised by the occurrence of MDs. In the case of more than 1% O_2, a profound filamentary discharge is observed [3]. Furthermore, the filamentary mode itself is affected by the oxygen content. For low oxygen content, the MDs occur almost always at the same value of the phase of the applied voltage. The same effect of MD reproducibility dependence on the gas composition is clearly seen in the current oscillograms. Visually it is found out that for low oxygen content, the MDs appear to burn at a steady state position between the electrode tips, while for the higher oxygen concentration, more diffuse shining discharge is observed.

An increase in oxygen content is found to result in characteristic changes in the emission spectrum. In all cases, the 2[nd] positive system of N_2 (SPS) dominates the spectrum. For low oxygen concentrations (0.5 and 1%), a weak signal of the NO_γ-system is clearly seen, while for higher values (more than 10%), the 0-0 transition of the 1[st] negative system of nitrogen (FNS) is observed. In fig. 1 only a part of the spectrum, presenting three different vibritional transitions (v'→v''= 0→3, 1→4, 2→5) of the SPS is shown. The structure of the SPS depends on the oxygen content. In the vibrational distribution for pure nitrogen and 0.5 % O_2 the 0-3 transition is the most intensive band, which is in contrast to the corresponding Franck-Condon factors (FCF). These observed overpopulation of the v'=0-level suggest that indirect mechanisms (e.g. via N_2-metastable states) are dominating the excitation process. The structure of the SPS-spectrum for higher oxygen admixture is in better agreement with the FCFs, thus referring to direct electronic excitation.

The plots of spatio-temporally resolved intensity distributions of selected spectral bands (SPS and FNS) for several gas mixtures are presented in fig. 2. Qualitatively, no significant difference in the microdischarge behaviour can be found here. The discharge evolution is characterised by a cathode directed luminosity wave followed by a glow at the anode. The velocity of the cathode directed luminosity wave (ionisation wave) seems to be independent of the gas composition (see fig. 3). However the higher oxygen content, the faster the decay of the SPS-signal (figure 4). Assuming the relaxation of the $N_2(C)$ density to be caused by spontaneous radiation and collisional quenching, reasonable agreement between measured and calculated data can be obtained:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_0} + K_{N_2} n_{N_2} + K_{O_2} n_{O_2} \approx K_{N_2} n_{N_2} + K_{O_2} n_{O_2}$$

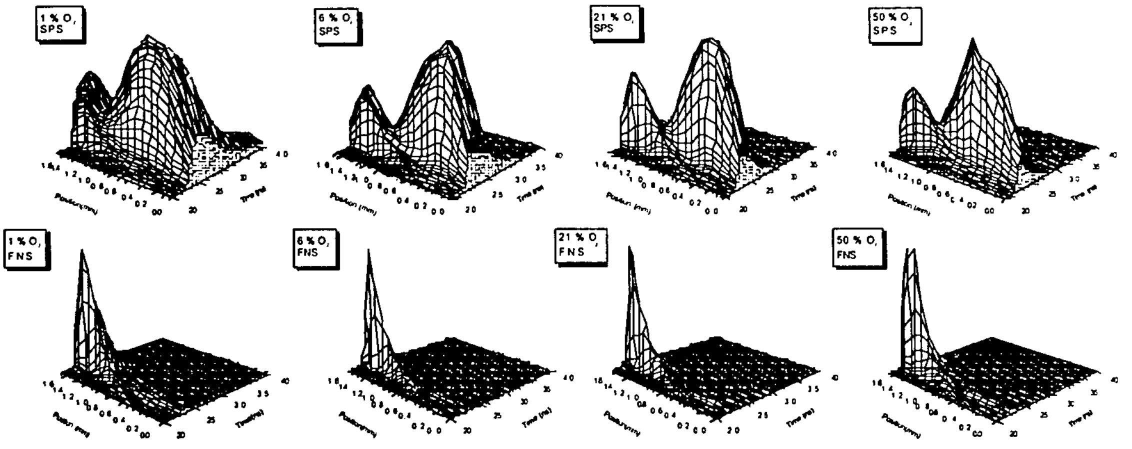

Figure 2: Spatio-temporally resolved intensity distributions for SPS and FNS, 0-0 transitions at λ = 337 nm and λ = 391 nm respectively (cathode at position 1.5 mm, anode at 0.1 mm)

where τ_0 is the radiative lifetime, K_X are the rate coefficients for de-excitation of $N_2(C)$ by X= N_2, O_2 taken from [4] and n_X are the densities of the species in the ground state. At atmospheric pressure the collisional quenching dominates. The best agreement between measured and calculated effective lifetimes is obtained in the case of stable microdischarge behaviour (3 vol. % O_2 admixture to N_2).

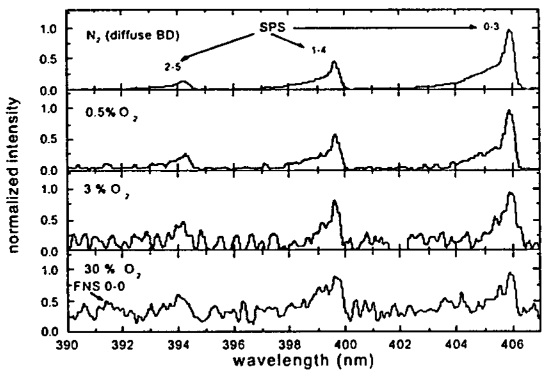

Figure 1: Comparison of emission spectra fragments for different gas mixtures

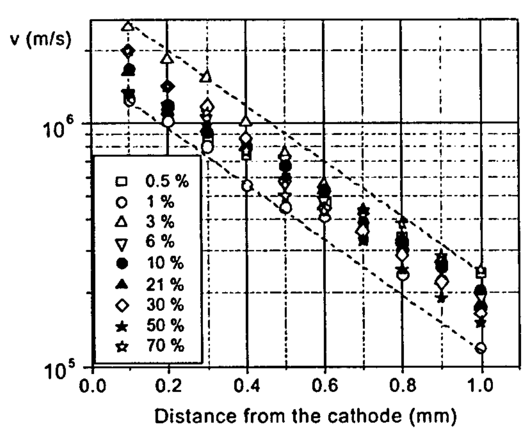

Figure 3: Velocity of the cathode directed wave as a function of axial co-ordinate and gas composition

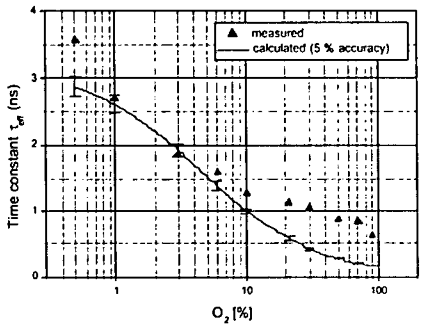

Figure 4: Effective lifetimes τ_{eff} for the SPS-signal

4. Summary

Qualitatively, no profound influence of the gas composition on the discharge behaviour has been found. Observed quantitative changes in the luminosity distributions depending on the oxygen content may be explained by the role of collisional quenching of nitrogen excited states by the molecules O_2.

5. Acknowledgements

The work was supported by Deutsche Forschungsgemeinschaft, SFB 198, "Kinetics of partially ionised plasmas".

6. References

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